SILICON CARBIDE RADIOISOTOPE BATTERIES

George C. Rybicki Photovoltaic Branch NASA Glenn Research Center Cleveland, Ohio 44135

ABSTRACT

The substantial radiation resistance and large bandgap of SiC semiconductor materials makes them an attractive candidate for application in a high efficiency, long life radioisotope battery. To evaluate their potential in this application, simulated batteries were constructed using SiC diodes and the alpha particle emitter Americium (241Am) or the beta particle emitter Promethium (147Pm). The 241Am based battery showed high initial power output and an initial conversion efficiency of approximately 16%, but the power output decayed 52% in 500 hours due to radiation damage. In contrast the 147Pm based battery showed a similar power output level and an initial conversion efficiency of approximately 0.6%, but no degradation was observed in 500 hours. However, the 147Pm battery required approximately 1000 times the particle fluence as the 241Am battery to achieve a similar power output. The advantages and disadvantages of each type of battery and suggestions for future improvements will be discussed.

INTRODUCTION

The maturation of new wide bandgap, highly radiation resistant semiconductor materials has made it worthwhile to revisit the topic of radioisotope batteries. SiC has a very desirable combination of properties for radioisotope battery design, including a large bandgap of 3 eV in the 6H polytype and a radiation induced atomic displacement threshold second only to diamond.(1) These attributes may make it possible to use an inexpensive radioisotopes to construct high efficiency, long lifetime radioisotope batteries. These batteries are potential power sources for applications where neither solar nor chemical battery power are feasible.

The operation of the SiC radioisotope battery is analogous to that of a solar cell except that high energy electrons or alpha particles are impinging on the cell rather than solar photons. Early radioisotope battery designs were based on beta emitting isotopes such as ¹⁴⁷ Pm and silicon diodes. (2) However, their performance was limited by the leakage current in the low bandgap Si diodes and the poor radiation resistance of Si. (3) Highly radiation resistant SiC semiconductor materials and diodes with very low leakage currents may make large performance increases in radioisotope batteries possible.

Previous attempts to use alpha particle emitting isotopes to fabricate a radioisotope battery have been made. (4) In that work, SiC photodetector diodes were irradiated with 5.5 MeV alpha particles from the radioisotope ²⁴¹Am. This isotope is inexpensive, widely available, and has a long half life of 458 years. It was also desirable because the alpha particles have a limited range in the semiconductor and this gives a better chance of capturing the generated carriers. The initial performance of the alpha particle based battery was impressive, but the power output rapidly decayed due to the high rate of radiation damage in the SiC diode materials.

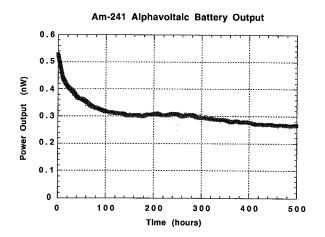
The advantages of SiC as an energy conversion material may still be realized in a betavoltaic energy conversion scheme. In this case, a beta radioisotope based SiC battery was constructed using the isotope ¹⁴⁷Pm. ¹⁴⁷Pm decays with the emission of high energy electrons, with an average energy of 62 KeV, and maximum energy of 225 KeV. Although the range of the beta particles is larger than that of the alpha particles, and thus the generated carriers may be harder to collect, the reduced radiation damage rate may make this design preferable.

EXPERIMENTAL

A simulated radioisotope battery was constructed by mounting a 2mm x 2mm SiC diode on a transistor header and placing it in a holder directly above the radioisotope source; this was a plated metal film source in the case of the 241 Am, and a Pm_2O_3 sealed disk source in the case of the 147 Pm. The diodes used in this study were SiC photodetector diodes obtained from the Cree Corporation in Durham, NC. Details of the structure and characteristics of the diodes are described elsewhere, (5) but briefly, they are composed of a p-type SiC substrate with an aluminum doped p-type epitaxial layer and nitrogen doped n-type emitter. The diodes were analyzed by the capacitance voltage technique before irradiation. A carrier concentration in the base of (2.25 \pm 0.6) x 1016 cm-3 was determined. The samples were then irradiated under vacuum with 5.5 MeV alpha particles using a 1.5 mCi 241 Am radioisotope source or with beta particles from a 30 mCi 147 Pm source. During the irradiation, the power output of the SiC cell was monitored as a function of time using an electrometer.

RESULTS

The power output in nanowatts as a function of time for the alpha and betavoltaic batteries are shown in Figure 1, Figure 2, and Table 1 below.



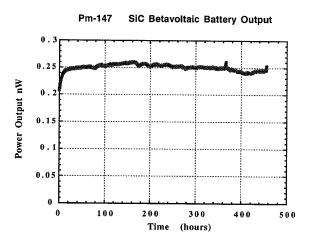


Figure 1. Alphavoltaic battery output

Figure 2. Betavoltaic battery output

Time (hours)	alpha battery power output	beta battery power output
0	0.527 nW	0.250 nW
100	0.321 nW	0.251 nW
200	0.304 nW	0.253 nW
300	0.298 nW	0.249 nW
400	0.276 nW	0.242 nW
500	0.259 nW	0.254 nW

Table I: Power output of alpha and betavoltaic batteries in nW as a function of time.

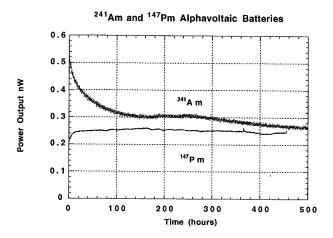


Figure 3. Output of Alphavoltaic and Betavoltaic batteries

The results clearly show that significant degradation occurs in the case of the alphavoltaic battery, but that essentially no degradation occurs in the case of the betavoltaic battery. The initial power output of the ²⁴¹AM battery degraded by 40% in the first 100 hours, 52% in the first 500 hours, and continued to decay at a lower rate, through 1000 hours and beyond. The comparative performance of the two batteries is illustrated more clearly in Figure 3. It is clear that the alphavoltaic battery had a higher initial power output, but after 100 hours the power output of the two batteries is comparable. However, the power output of the two cells should be considered in terms of the strengths of the isotope and their geometric configurations. The fluence at the sample surface was 6000 alpha particles per second per mm², in the case of the alpha battery, and was 5.8 X 10⁶ electrons per second per mm2 in the case of the beta battery. The net result was that it took approximately 1000 times as much beta flux as alpha flux to generate a similar amount of power. Part of this is certainly due to the fact that the beta particles had much less energy than the 5.5 MeV alpha particles, (62 KeV on average and 225 KeV maximum). Based on the 62 KeV average energy, the beta particles had on 90 times less energy than the alpha particles, and would thus generate fewer electron hole pairs. The remaining difference, accounting for the required higher beta fluence, is likely related to the longer range and thus greater difficulty in collecting the electron hole pairs generated by the beta particles. The 5.5 MeV alpha particles have a range of approximately 28 μm in SiC, but the beta particles have a range of up to 300 μm. The diffusion length in SiC is only a few microns (6) so the longer particle range makes it more difficult to collect the energy of the beta particles.

The effect of the particle range on energy collection can be demonstrated by making an estimate of the power conversion efficiency of the two simulated batteries. A commonly used theoretical model predicts that the energy lost in the creation of an electron hole pair in an absorber by a charged particle is $2.8 E_g + 0.5 \text{ eV}$, or 8.9 eV in SiC. (3) Each alpha particle could thus generate 616,000 electron hole pairs and each beta particle, on average, 6965 electron hole pairs. At the measured fluxes in these experiments, this would result in the generation of 1.48×10^{10} carriers per second or 2.4 nA in the case of the alphavoltaic battery and 1.6×10^{11} carriers per second, and 25.8 nA in the case of the betavoltaic battery. The actual collected currents were 0.382 nA in the case of the alphavoltaic battery, or a 16% conversion efficiency, and 0.148 nA in the case of the betavoltaic battery, or 0.6% conversion efficiency. In this case the advantage of the alphavoltaic approach is clear.

In a previous study, the degradation of the power output of the simulated alphavoltaic battery was correlated with radiation damage in the diodes, and specifically to the introduction of deep levels in the SiC diodes. (4) The introduction rate of these defects was also measured under both 1 MeV electron irradiation and 5.5 MeV alpha particle irradiation. Of course, the introduction rate of defects during electron irradiation was much lower than that in the case of alpha particle irradiation; a defect introduction rate 5700 times lower was measured for the 1 MeV electrons. If the collision stopping power of 1 MeV electrons is compared to 62 KeV and 225 KeV electrons, the production of defects by the lower energy electrons might be expected to be lower still.

The degradation of battery output under electron irradiation is not observed here, probably as a result of a combination of effects. First, it would require several thousand times as much fluence under beta particle irradiation as alpha particle irradiation to generate a equivalent number of defects. Another factor that may contribute is that the minimum energy required to displace an atom in SiC, at 21.8 eV, is higher than in almost any other material, and results in a displacement threshold of 108 KeV in SiC (1). The electron energy spectra for the beta decay of ¹⁴⁷Pm has an average energy of 62 KeV, which is below the threshold energy for lattice damage in SiC. For these reasons, no power output degradation was observed in the betavoltaic battery, and in the absence of any apparent degradation in power output, no studies were made of deep levels introduced by the ¹⁴⁷Pm irradiation.

The implications of these results is that similar power outputs can be obtained using alpha particle emitters or properly selected beta particle emitters in the construction of semiconductor based radioisotope batteries. The advantage of the higher energy, shorter range alpha particles is reduced by the higher rate of radiation damage they cause. Several attempts were made to reduce the rate of radiation damage in the alpha particle based radioisotope battery, for example thicker emitters, larger surface area diodes and junction edge protection were all investigated. Unfortunately none of these approaches significantly reduced the radiation damage rate. (4)

Several other avenues may still be available for the improvement of these radioisotope batteries. For example, lower energy alpha emitters and higher and lower energy beta emitters should be investigated. For the isotope selected, both the energy of the decay product and the half life must be considered. A very long half life results in a low specific activity and an impractical amount of radioisotope material would be required. A very short half life would make the battery impractical to use before its output decayed. Alpha isotopes are suggested that had reasonable half lives and alpha energies significantly less than 5.5 MeV. Beta isotopes were suggested that had an average decay energy of less 108 KeV. A table of candidate isotopes is presented below.

Material	Half Life (yrs)	Alpha Energy (MeV)	Beta energy (KeV)	
¹⁴⁸ Gd	74.6	3.183		
63 N i	100		66 max,	17.1 avg
14C	5730		156 max	49.5 avg
₆₀ Co	5.27		317 max	95 avg
⁹⁹ Tc	21300		293 max	85 avg
241Am	432	5.49		
¹⁴⁷ Pm	2.63		225 max	62 avg

Table 2: Potentially useful isotopes for radioisotope batteries

Although there are hundreds of available radioisotopes, few are suitable for these applications. Even the use of those isotopes thought to be suitable may be complicated by radiation hazards as in ⁶⁰Co, or high cost and limited availability as in ⁶³Ni. The use of ¹⁴⁸Gd in the alpha battery may look promising as it is a lower energy alpha emitter than ²⁴¹Am, but the alpha particle energy still far exceeds the damage threshold in SiC. In the case of other potential beta emitting isotopes, ⁶⁰Co looks nearly ideal but its use might be complicated by the strong gamma emission associated with its decay. Isotopes such as ²⁰⁴Tl have been suggested, but they have an average electron energy higher than the damage threshold in SiC. Others such as tritium or ¹⁴C have energies less than that of ¹⁴⁷Pm and would produce less electron hole pairs. However, low energy beta emitting isotopes may have some potential, as they could result in higher conversion efficiencies due to the shorter range of the electrons in the diode material.

Conclusions

Both alpha and beta particle emitting radioisotopes can be used to construct semiconductor based radioisotope batteries. The alpha based design shows high efficiency and power output, but a shorter lifetime due to the severe radiation damage caused by the alpha particle irradiation. The beta particle based design, on the other hand, showed lower efficiency but no degradation over time. The beta design could probably be loaded with more isotopes to compensate for its lower efficiency, but no easy solution appears to be available for the radiation damage problem with the alpha based design. Additionally, experiments with low energy, short range beta emitters may result in a more efficient, higher output betavoltaic battery design.

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